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Estimation of the contribution of a municipal waste incinerator to the overall emission and human intake of PCBs in Wilrijk, Flanders

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Abstract

The contribution of the emission of PCBs by a municipal waste incinerator in Wilrijk, a relatively industrialized district in the largest city of Flanders, to the total emission to air and to the total human intake was estimated. Therefore it was compared to the emission of PCBs by evaporation from PCB containing applications (transformers, capacitors, paint, ink, etc.) and to the intake of PCBs with food. As there was a lack of PCB data from the incinerator, the PCB emission concentration was estimated using three different approaches. A PCB measurement of the incinerator emission, performed later on, fell within the predicted range of 0.0004–0.005 ng TEQ/Nm^3.

Emission of PCBs from PCB containing applications and intake from food were deduced from information available on Flemish and European level. The results indicate a PCB contribution from the incinerator to local emissions between 0.3% and 3% of the emission from PCB containing applications and a contribution to human intake less than 6×10^{-4}% of the intake from food.

Keywords: Polychlorinated biphenyls (PCBs); Waste incineration; Food contamination; Transformers; Capacitors; Emissions

1. Introduction

The United States Environment Protection Agency (EPA) and Environment Canada (1999) identified several sources of PCBs to the environment:

- evaporation and leaks from PCB containing applications. PCBs have been used in closed and open applications. Closed applications include transformers, capacitors and other electrical components such as switches, pressure regulators and circuit breakers. Small capacitors, ink, paint, oil, plasticizer and adhesive are examples of open applications. A EU Directive (96/59/EG, 1996) aims at cleaning and/or destroying all closed applications by the end of 2010. Flanders has worked out a destruction plan from 2000 until 2005, with possible exceptions until 2010;
- incineration of waste materials containing or not containing PCBs;
- chemical processes involving carbon, chlorine and high temperature;
- illegal dumping of PCB containing waste and bad management of PCB contaminated landfills or sites.

Because of their resistance against biodegradation and their hydrophobic properties, PCBs are mainly found in air, soil and sediments and less in water, and

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accumulate in fat in the food chain (Fiedler, 1997; ATSDR, 2000).

In Flanders, few measured data are available on PCB sources and concentrations. The PCB concentration in emissions is not regulated and is therefore seldom measured, contrary to polychlorinated dibenzo-*p*-dioxin and dibenzo furans (PCDD/PCDF).

Wilrijk is a district of Antwerp, the largest city in Flanders, and has about 40 000 inhabitants. A municipal waste incinerator, which may contribute to PCB emission and human intake, is situated in Wilrijk. In a recent investigation of the Flemish government on the link between environment and health it was implicitly expected to find higher concentrations of PCBs in human blood samples due to the presence of, among others, the incinerator. As a consequence government agencies requested monitoring of PCB emission from this incinerator.

In this context we wanted to investigate whether emissions from the incinerator can be expected to contribute significantly to the overall emission and human intake of PCBs in Wilrijk. Therefore we estimate the emission of PCBs by the municipal waste incinerator in Wilrijk and compare it to the emission of PCBs by evaporation from open and closed applications. Moreover we compare the total human intake of PCBs from the incinerator by respiration to the intake of PCBs from food. Initially no actual measurements of the PCB emissions were available. So we estimated these emissions from available data. In a later stage, measurements became available.

Estimates should take into account the relative toxicity of the different PCB congeners. Ogura et al. (1999) showed that dioxin-like PCBs contribute for less than 10% to the total TEQ in ambient air, but that sometimes this contribution can increase to 80% possibly due to the presence of PCB containing applications and to combustion of particular types of waste. Sakai et al. (1999) showed for a particular incinerator in Kyoto that although PCBs were decomposed in the incineration process, the emission output in TEQ was higher than the input. This conclusion was however contradicted by findings of Sakai et al. (2001) for another, more recently constructed, municipal solid waste incinerator where the emission of all PCB congeners with a TEF value was lower than the input. This study also demonstrated the use of congener profiles from deposition samples to evaluate PCB sources. The present study will however only focus on total TEQ for the evaluation of the contribution from a source.

2. Estimation of municipal waste incinerator emission

We adopted three approaches to estimate the PCB emission concentration for the Wilrijk incinerator from available information:

- based on PCB emission concentrations of municipal waste incinerators in Japan and Wallonia, i.e. the southern part of Belgium, with similar flue gas cleaning technology;
- assuming that the average ratio PCDD–DF/PCB in the stack emissions of incinerators in Japan and Wallonia equals the one of the incinerator in Wilrijk, and using the measured PCDD–DF emission concentrations of the Wilrijk incinerator;
- assuming that the ratio PCDD–DF/PCB in fly ash and air pollution control residue, for which there are measurements in Wilrijk, equals the one in the stack emissions.

2.1. Comparing with the waste incinerators in Japan and Wallonia

The PCDD/PCDF emission concentration in the stack gas of the Wilrijk incinerator is frequently measured and is on average 0.02 ng TEQ/Nm^3. Table 1 gives dioxin and PCB emission concentrations from two municipal waste incinerators in Kyoto, Japan (Sakai et al., 1999; Sakai et al., 2001), and from four incinerators in Wallonia, Belgium (Wallonie, 2001). The flue gas cleaning technology in these incinerators is shown in Table 2.

As can be seen from the PCDD/F emission and the cleaning technology, Charleroi is an old incinerator with low quality flue gas cleaning. Liege and Kyoto1 have also a limited cleaning technology. Virginal, Thumaide and Kyoto2 are the most recent installations and compare best to the installation in Wilrijk. Since the measurement campaign in 1999, air pollution control technology in the Walloon incinerators was seriously improved.

The PCB concentration of 0.005 ng TEQ/Nm^3 in Virginal will be considered as an upper limit for the emission in Wilrijk.

2.2. Using the PCDD–DF/PCB ratio in the stack emissions

The ratio PCDD–DF/PCB in the stack emissions (Table 1) appears relatively similar for the six considered incinerators (Charleroi, Liege, Thumaide, Virginal and Kyoto1 and 2), if two very low values (1.1 and 7.5) for the incinerator in Charleroi, which is not state-of-the-art, are not considered. A mean ratio of 45 (s.d. = 25) is obtained ($n = 9$).

With this ratio, we obtain a PCB emission concentration of 0.0004 ng TEQ/Nm^3 derived from the measured PCDD/F concentration of 0.02 ng TEQ/Nm^3.

2.3. Using the PCDD–DF/PCB ratio in fly ash and air pollution control residues

Measurements of PCDD–DF and PCBs in the fly ash from the ESP, in the baghouse ash and in the acid and

Table 1
Emission concentrations (mean of three measurements) at the time of a measurement campaign in 1999 in Wallonia, together with data for Wilrijk and Kyoto

	Oven line no.	PCDD/F emission concentration[a] (ng TEQ/Nm^3)	PCB emission concentration[b] (ng TEQ/Nm^3)	Ratio PCDD–DF/PCB
Wilrijk	2	0.02	–	–
Charleroi	1	229	30	7.5
	2	99	89	1.1
	3	23	0.6	40
Liege	1	2.3	0.02	97
	2	4.4	0.11	40
	3	4.3	0.08	56
	4	2.7	0.10	28
Thumaide	1	0.75	<0.03	–
	2	0.25	<0.035	–
	3	0.12	<0.01	–
Virginal	1	0.12	0.005	23
Kyoto1	1	14	0.23	61
	2	1.5	0.03	50
Kyoto2	–	0.036	0.0031	12

[a] Sum of 17 PCDD/PCDF that have a TEF.
[b] For the Walloon incinerators, sum of PCB 77, 126 and 169, the three PCBs with the highest TEF value. For Kyoto, the sum of all PCBs that have a WHO–TEF value.

Table 2
Flue gas cleaning technology at the time of a measurement campaign in 1999 in Wallonia, together with data for Wilrijk and Kyoto

Incinerator	Flue gas cleaning
Wilrijk	Non-catalytic deNO_x; ESP; semi-wet injection of lime slurry and activated carbon; baghouse filter; wet two stage scrubber
Charleroi	Wet scrubber
Liege	Dry injection of lime; ESP
Thumaide	Semi-wet injection of lime slurry and activated carbon; baghouse filter; wet two stage scrubber
Virginal	Semi-wet injection of lime slurry; ESP; injection of lime and activated carbon
Kyoto1	Dry injection of lime; ESP
Kyoto2	Baghouse filter; dry injection of slaked lime; catalytic denitrification

basic scrubber residue from the Wilrijk incinerator were available in Buekens and Segers (2000) and are shown in Table 3. The mean PCDD–DF/PCB ratio in the considered incinerator residues is 4.6 (s.d. $= 3.3$; $n = 4$).

Assuming this ratio for the stack emissions, a PCB emission concentration of 0.0044 ng/Nm^3 is obtained from the measured PCDD/F concentration.

2.4. Comparison of the three calculation approaches and estimation of PCB emission

The three approaches lead to an emission concentration of 0.0004–0.005 ng TEQ/Nm^3. With an emission flow rate of 102 000 Nm^3/h, a PCB emission of 45–510 ng TEQ/h or 0.39–4.5 mg TEQ/y is obtained. The 1 order of magnitude spread between lower and upper estimate seems acceptable for our purpose.

The validity of this approach was supported later on by a measurement at the Wilrijk incinerator (Adams, 2001). A congener-specific analysis of the emission in the stack gave a PCB emission concentration of 0.00146 ng TEQ/Nm^3, falling within the estimated range. As the measurement reported is only a single measurement, we will proceed with the estimated interval to evaluate contributions.

In the next paragraphs we have opted for a worst-case scenario (ambient air concentration in the point of highest concentration, respiratory intake in the point of highest impact) in order to overestimate the possible contribution of the incinerator.

Table 3
PCDD, PCDF and PCB concentration in fly ash and air pollution control residue of the Wilrijk incinerator

	PCDD (ng TEQ/g)	PCDF (ng TEQ/g)	PCB (ng TEQ/g)	PCDD–DF/PCB ratio
Fly ash ESP	162	87.1	27.6	9.0
Baghouse ash	40.2	90.8	28.6	4.6
Acid scrubber	1290	702	1492	1.3
Basic scrubber	2564	1773	1287	3.4

3. Immission from the municipal waste incinerator in air and human intake by respiration

The TA-Luft dispersion model (BMU, 2002) was used to calculate yearly averages of immission concentration in the air from the incinerator emission. Based on meteorological data over 38 years, the frequency of occurrence of a certain atmospheric situation (determined by 36 possible wind directions, 6 stability classes and 9 wind velocities) is calculated. A peak ambient air concentration of 0.0028–0.032 fg TEQ/Nm^3 is found at 700 m NE from the stack of the municipal waste incinerator.

Assuming a daily intake of 22 m^3 of air by an average person of 65 kg, a maximum PCB intake by respiration of 0.062–0.71 fg TEQ/day is obtained.

4. Emission from open and closed applications

OVAM, the Flemish Waste Agency, manages a database concerning the presence in Flanders of closed applications. The main closed applications are transformers and capacitors, representing more than 99.5% of the total amount of PCBs in closed applications. The information for Wilrijk was obtained from this database. Table 4 gives the situation in Wilrijk in the beginning of 2001, together with specific emission factors for transformers and capacitors (Annema et al., 1995; FEA, 1998).

In 2000, the amount of PCBs still present in open applications in Belgium was estimated, together with the related release (OSPAR, 2001). A release of 5.52 ton of PCB was found. This amount however is the sum of release to air, water and soil from small capacitors (3.15 ton) and evaporation to air from other open applications (2.37 ton). The emission to air from the small capacitors is estimated to be 10% of the total release (Annema et al., 1995). The emission of open applications is then 2.69 ton. With a population ratio Wilrijk/Belgium of 38 138/10 239 085 (January 1, 2001) this emission is recalculated to be 10.0 kg.

These emissions have to be transformed into TEQ. However, no information is available on the relative amounts of congeners in the PCBs used in the open and closed applications. In Flanders the most used brand of PCB fluids was Aroclor. The main types were Aroclor 1260 (60% chlorine), Aroclor 1254 (54% chlorine), Aroclor 1242 (42% chlorine) and Aroclor 1016 (41% chlorine). Using the fingerprints of these Aroclor types (Schulz et al., 1989) and assuming that the four types were used in equal amounts, the TEQ of 1 kg of Aroclor is calculated to be 9.975 mg PCB (Table 5).

Table 6 gives the total annual PCB emission of closed and open applications. The calculated emission of 143 mg TEQ/y from open and closed applications is more than 30 times the emission estimate from the Wilrijk incinerator.

Table 4
Closed applications of PCBs in Wilrijk (based on an inventory of amount on February 28, 2001) and emission factors

	PCB fluid (kg)	Emission factor (%/y)
Transformers	38 559	0.006
Capacitors	1260	0.16

Table 5
TEQ value of 1 kg of Aroclor type

Aroclor type	mg TEQ PCB/kg product
Aroclor 1016	0.0
Aroclor 1242	5.3
Aroclor 1254	19.5
Aroclor 1260	15.1
Mean (each 25% in use)	9.975

Table 6
PCB emission from PCB containing applications in Wilrijk

	PCB emission (kg/y)	PCB emission (mg TEQ/y)[a]
Closed applications		
Transformers	2.3	23
Capacitors	2.0	20
Open applications	10.0	100
Total	14.3	143

[a] This value is calculated considering the TEQ value per kg Aroclor in Table 5.

The calculated emission from open and closed applications relies on the quality of the inventory of closed applications, the used emission factors and the estimate of emission from open applications. The amount of PCB fluids in closed applications is an underestimation because not all weights of PCB fluids in the small applications are known. Emission factors are a major uncertainty but the quoted factors are used in most estimations of emissions from open and closed applications (Annema et al., 1995; FEA, 1998; OSPAR, 2001).

The original estimate from open applications was carried out for Belgium. Using the population ratio Wilrijk/Belgium will again underestimate the emission for Wilrijk, because this ratio takes into account only the use of open applications by the population and not by industry, which is highly present in Wilrijk. We opted for these underestimations, again in order to overestimate the contribution of waste incineration. A major uncertainty however, and thus maybe an overestimation, is the relative presence of the different Aroclor types in open and closed applications. We opted for an equal use of the four main types, but this choice greatly influences the result because of their diverging TEQ values (Table 5). Over the years less chlorinated Aroclors were produced, but many of the old Aroclor fluids are still present in older devices. Even when only Aroclor 1016 and 1242 are supposed to be present in equal amounts, the total PCB emission from open and closed applications will be 38 mg TEQ/y, about ten times higher than the contribution from the incinerator.

Since closed applications using PCB fluids have to be destroyed by 2005 (exceptions possible until 2010), this source will disappear. PCB emission from open applications will also gradually decrease to zero. As a consequence, the relative contribution of other sources will increase.

5. Human intake by means of food

As PCBs accumulate through the food chain, food is a major contributor to human intake. A report of the European Commission (Anonymous, 2000) estimated the daily intake of PCBs through food to be 0.8–1.8 pg TEQ/kg body weight. According to the report, this would represent more than 90% of total intake. Our own calculations based on the European per capita consumption (WHO, 1998), the fat content of the different food products (Nubel, 1992) and the PCB concentrations in the different food products (Anonymous, 2000), result in a daily intake of 1.79 pg TEQ/kg body weight. This equals 116.3 pg TEQ for an average person of 65 kg. The largest contribution comes from fish (34%), milk and dairy products (25.5%) and meat (17.5%).

Possible sources of food contamination include the deposition of PCB emissions on vegetation and respiration of PCBs by animals, contaminated animal feed, contamination during the food production, use of contaminated sludge as a fertilizer and contaminated soil and surface water. The contributions to food contamination of emissions from PCB containing applications and waste incinerators are not included in the previous calculations because the inhabitant of Wilrijk is consuming food not necessarily originating from Wilrijk. Sometimes vegetables grown in the kitchen garden will complement the meal but they hardly contain PCBs (no fat) and as they are usually thoroughly washed, the impact of deposition of PCBs will be lessened.

The human intake of PCBs by means of food is well documented and the reliability is considered to be good.

6. Evaluation of the contributions

Table 7 gives an overview of all the obtained results. The PCB contribution of the Wilrijk incinerator is compared to the emission from PCB containing applications and to the human intake with food.

Although the estimated emission concentrations of the Wilrijk incinerator differ by 1 order of magnitude, the accuracy of the interval is considered sufficient, in view of the three approaches that were used to obtain the results. A subsequent actual PCB measurement fell within the interval.

The estimation was performed in a worst-case scenario for the incinerator, comparing the maximum immission and respiration contributions for the incinerator to conservative calculations for other sources. The incinerator appears to contribute to local emissions 0.3–3% of the emission from PCB containing applications.

Table 7
Overview of PCB contributions of different sources

	Emission from Wilrijk incinerator	Emission from PCB containing applications
PCB emission to air (mg TEQ/y)	0.39–4.5	143
	Respiration of PCBs from Wilrijk incinerator emission	Eating of food
Human intake (fg TEQ/day)	0.062–0.71	116 300

To human intake the incinerator contributes between 5×10^{-5} and 6×10^{-4}% of the intake from food.

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